

# Electron-mediated ferromagnetism and small spin-orbit interaction in a molecular-beam-epitaxy grown n-type $GaAs/Al_{0.3}Ga_{0.7}As$ heterostructure with Mn $\delta$ -doping

A. Bove,<sup>1,2</sup> F. Altomare,<sup>3</sup> N. B. Kundtz,<sup>1</sup> A. M. Chang,<sup>1,\*</sup> Y. J. Cho,<sup>4</sup> X. Liu,<sup>4</sup> and J. Furdyna<sup>4</sup>

<sup>1</sup>*Physics Department, Duke University, Durham, NC 27708*

<sup>2</sup>*Physics Department, Purdue University, West Lafayette, IN 47906*

<sup>3</sup>*NIST, 325 Broadway, Boulder, Colorado 80305*

<sup>4</sup>*Department of Physics, University of Notre Dame, Notre Dame, Indiana 46556*

(Dated: July 2, 2008)

We report on transport measurements indicating electron-mediated ferromagnetism in a molecular-beam-epitaxy (MBE) grown  $GaAs/Al_{0.3}Ga_{0.7}As$  heterostructure with Mn  $\delta$ -doping. The interaction between the magnetic dopants (Mn) and the Two-Dimensional Electron Gas (2DEG) gives rise to magnetic ordering at temperatures below the Curie temperature ( $T_C \sim 1.7K$ ) when the 2DEG is brought in close proximity to the Mn layer by gating. The Anomalous Hall Effect (AHE) contribution to the total Hall resistance is shown to be three orders of magnitude smaller than in the case of hole-mediated ferromagnetism indicating the near absence of spin-orbit interaction.

PACS numbers: 75.47.-m, 75.50.Pp, 85.35.Be

In diluted magnetic semiconductor (DMS) systems, when a high concentration of magnetic impurities is incorporated in the non-magnetic semiconductor hosts, ferromagnetic behavior is observed. In these systems, magnetism is produced by a carrier-mediated interaction between magnetic impurities. In DMS systems such as the promising ferromagnetic GaMnAs systems, the origin of ferromagnetism has been proven to be hole-mediated ([1], [2]).

To date, the only possibilities for n-type, electron-mediated ferromagnetism are found in the large bandgap GaMnN system ([3],[4]) and the ZnMnAlO system [5]. Even though evidence of ferromagnetism is observable at temperatures above room temperature, due to a scarcity of magneto-transport data, the claim that magnetism is carrier-mediated rather than originating from the clustering of the Mn atoms has remained controversial.

In this letter, we present direct evidence in magneto-transport measurements demonstrating electron-mediated ferromagnetism in a specially designed, low carrier concentration ( $\sim 10^{12}cm^{-2}$ ) GaMnAs quantum well (QW). In our structure, the ferromagnetism is controllable via tuning of the n-carrier density. The ferromagnetism is manifested in unambiguous, hysteretic behavior of the transport coefficients under the application of a magnetic field. Hysteresis is present only when the GaMnAs quantum well is filled with electrons via gating, and is absent when the electrons only occupy a heterojunction (HJ) situated far away from the Mn atoms. Moreover, in our system, there is a clear absence of the anomalous Hall effect (AHE) -an effect rising from spin-orbit scattering in the presence of a finite magnetization. This absence stands in contrast to what has been widely observed in all other GaMnAs systems, which were all p-type systems, and is consistent with a GaAs conduction band, which is primarily s-band in

character.

Our n-type crystal containing a  $\delta$ -doping Mn layer in a GaAs QW was grown by molecular-beam-epitaxy on a semi-insulating (SI) GaAs (001) substrate (see table 1 for the growth structure). The 2/3 monolayer (ML) of Mn was grown at the GaAs/AlGaAs interface situated closer to the crystal top surface while keeping the substrate at low temperature (LT)  $\sim 250^\circ C$ .

TABLE I: Heterostructure

GaAs (001)	Substrate
GaAs:Si	100 nm
AlGaAs:Be	200 nm
SI-GaAs	10 nm
LT-Mn	$\frac{2}{3}$ ML
LT-AlGaAs	10 nm
LT-AlGaAs:Be	15 nm
LT-GaAs	5 nm

Based on previous studies ([10], [11]), at such low growth temperature the Mn atom should be free of clustering, and are uniformly distributed. In this structure, a layer of 2D electron gas (2DEG) is always present at the interface of the Si doped 200 nm GaAs and the 100 nm AlGaAs:Si/Be layers,  $\sim 140$  nm below the surface, while the 10 nm GaMnAs QW can be filled or emptied by gating.

To fabricate devices for transport measurements, it is necessary to make good ohmic contact. A novel method was developed for annealing ohmic contact, which employs a thermal gradient along the sample. This method is able to preserve the ferromagnetism in the center of the sample, where the Hall bar is located, while destroying it in the vicinity of the contacts [6]. Following annealing of the contacts, standard lithographically and wet-etching were performed to define a Hall bar with a 150

$\mu\text{m}$  wide channel. The voltage probes were narrow, 10  $\mu\text{m}$  in width, to minimize their perturbation on the current flow. Magneto-transport measurements were carried out using a lock-in amplifier at 3.7 Hz and 10 nA excitation current.

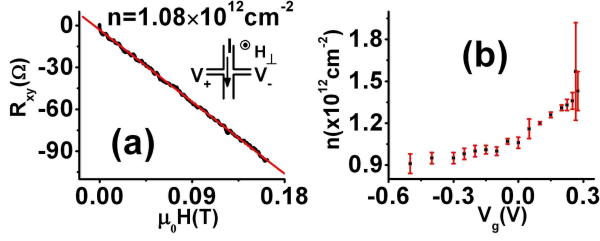


FIG. 1: (a) Carrier density of the 2DEG at  $T=4.2\text{K}$ . (b) Gate voltage dependence of the carrier density (Note the change in slope near  $\sim 0\text{V}$ , indicating filling of the GaMnAs QW). Note that the  $R_{xy}$  was anti-symmetrized with respect to  $H$  to remove residual contributions from  $R_{xx}$

The first task is to establish the carrier type. The sign of the Hall resistance ( $R_{xy}$ ) in Fig. 1(a) indicates that our structure contains n-type carriers. An estimate of the carrier density using a one component carrier model to analyze the Hall coefficient ( $n \propto 1/R_{Hall}$  rather than a two component model, one in the QW and the other at the Si-doped GaAs and AlGaAs:Si/Be interface) yielded a carrier density  $n \sim 1.08 \times 10^{12} \text{cm}^{-2}$ , and a mobility  $\mu \sim 575 \text{cm}^2/\text{Vs}$  at 4.2K [6]. This density has a sizable error of  $\sim 50\%$  arising from the assumption of one-component [6], but gives a rough characterization when the mobility and density in the two components are roughly comparable, as is the case here. To further establish that the carrier type inside the QW is the electron, the gating dependence of the  $R_{xy}$  was examined after deposition of a metal gate. The resultant carrier density is shown in Fig. 1(b) as a function of the applied voltage. A more positive gate voltage increases the density, clearly establishing the carrier type to be n-type. Notably, there is an abrupt change in slope near  $V_g \sim 0\text{V}$ , indicating the filling of the GaMnAs quantum well (QW) for positive gate voltages. The slopes for  $V_g > 0\text{V}$  and  $V_g < 0\text{V}$ , yield capacitances corresponding to a 2DEG distance of  $\sim 50 \pm 10\text{nm}$  and  $\sim 210 \pm 30\text{nm}$ , compared to the nominal distances of 35 nm and 145 nm, respectively [6]. The distances, 40% larger than nominal, could result in part from a higher crystal growth rate than nominal, which was calibrated to  $\sim 25\%$  accuracy.

To investigate the magnetism, our device was cooled to 0.3 K and an in-plane magnetic field  $H_{||}$  was applied. At zero gate voltage, no hysteresis was observed in  $R_{xy}$  (black traces in Fig. 2(a)). However, after applying a positive gate voltage of 500 mV to fill the GaMnAs QW with electrons, a clear hysteretic loop appeared in the hall signal (red traces in Fig. 2(b)). Similar behavior was observed in several Hall junctions and in the longitudinal

resistance  $R_{xx}$ . At the same time,  $R_{xx}$  exhibited a small but reproducible feature as a function of temperature, indicative of a ferromagnetic transition near 1.7 K (Fig. 2(c)).

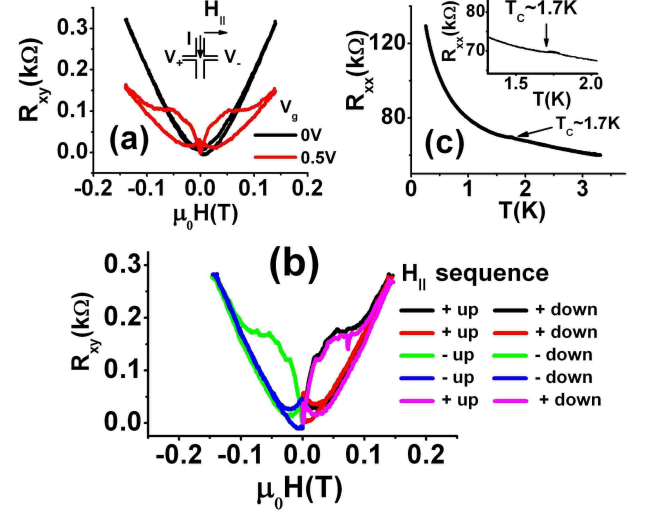


FIG. 2: (a) Hysteresis loops (red traces) are observed after a gate voltage of 0.5V is applied at  $T=0.3\text{K}$ . (b) History dependent behavior at  $T=0.3\text{K}$  with an in-plane magnetic field applied. When magnetism appears, the  $R_{xy}$  traces often no longer appear anti-symmetric in  $B$ , despite the well-defined Hall bar geometry. (c) Longitudinal resistance as a function of the temperature. The bump at around 1.7K is attributed to the ferromagnetic phase transition.

To further substantiate the hysteretic behavior, the magnetic field was swept up and down twice in the positive direction before changing its sign. When the magnetic field was first swept up and down (black traces), a clear loop appeared, while for the traces taken subsequently without reversing the magnetic field (red traces), the loop is absent. Similar behavior was observed for negative field traces (green and blue, respectively).

To elucidate the n-type mediated ferromagnetism, we studied the AHE and compared it with reported systems where ferromagnetism is known to be hole-mediated. To date, experimental studies of the AHE in electron-mediated systems are still lacking ([3], [4] and [5]), while there are only a few theoretical predictions ([7],[8]). As a result of a much reduced spin-orbit coupling, it is believed that the AHE should either be absent or much smaller than in hole-mediated systems [7].

For this purpose, a new device was prepared and measured the in a perpendicular field  $H_{\perp}$  versus temperature. The advantage of this second device resides in the fact that possibly due to either inhomogeneity in the local strain from the LT growth or a gradient in the Mn concentration, this second sample had areas that showed ferromagnetism while others did not as it can be seen in Figs. 3(a) and 3(b). Note that without strain or residual

dipole interaction to introduce anisotropy in the Heisenberg spin coupling between Mn impurities, there should, in principle, be no ferromagnetic state at a finite temperature for this 2D system.[12] We are thus able to study the AHE for a purely paramagnetic (PM) system (non-magnetic junction) and for another that shows a ferromagnetism (FM) (magnetic junction) when the temperature is below the Curie temperature  $\sim 2K$  (Fig. 4(a)).

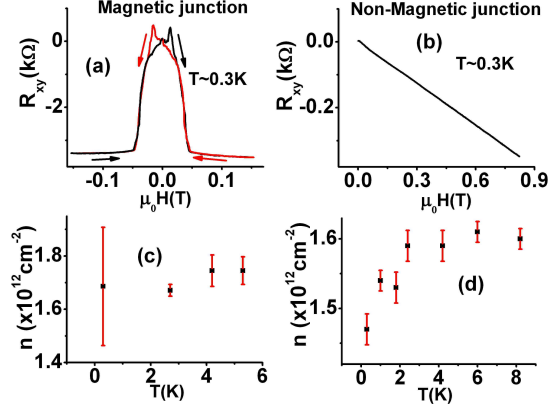


FIG. 3: (a) Hall resistance for the ferromagnetic junction. (b) Hall resistance for the paramagnetic junction. (c) Carrier's density for the ferromagnetic junction as a function of temperature. (d) Carrier's density for the paramagnetic junction as a function of temperature. The magnetic field is perpendicular to the sample's surface in all four cases.

The Hall resistance  $R_{xy}$  versus  $H_{\perp}$  was measured at different temperatures for both the FM and PM junction and the density deduced from  $n \propto 1/R_{Hall}$  are plotted in Figs. 3(c) and 3(d), respectively. A detailed two-component analysis of the Hall coefficient  $R_{Hall}$  is presented below. For the FM junction, the density (Fig. 3(c)) remained constant within experimental error. On the other hand, for the PM junction the density shown in Fig. 3(d) remained constant above 2 K, but decreased as the temperature was further reduced down to  $T \sim 0.3K$ . This latter behavior is similar to what is expected from an AHE contribution to the Hall resistivity ([2],[8]). Jungwirth et al. [8] have suggested that a true AHE should saturate at some high field value and the real Hall coefficient should be restored above such a field. To distinguish an AHE contribution from the magnetic freeze-out of carrier, we examined the Hall resistivity at higher magnetic fields, beyond the saturation of Mn spins. Lowering the temperature to  $T \sim 0.3K$  and sweeping the magnetic field up to  $\sim 0.8$  T, the  $R_{xy}$  did not show any appreciable non-linearity in that field range, indicating that the AHE contribution must be small.

To quantify the AHE coefficient, we fitted the  $R_{Hall}$  as a function of temperature for the PM junction using

the standard expression for the Hall resistivity  $\rho_{Hall}$ :

$$\rho_{Hall} = R_{Hall}\mu_o H = R_0\mu_o H + R_S M \quad (1)$$

where  $R_0$  is the Ordinary Hall Effect (OHE) coefficient,  $R_S$  is the AHE coefficient and  $M$  is the magnetization.  $M$  for a paramagnet is proportional to the magnetic susceptibility  $\chi$  that obeys the Curie-Weiss law  $\chi = C/(T - T_C)$ .  $C$  is the Curie constant determined in [9] and is theoretically expressed as  $C = (4p_{eff}^2 \mu_B^2 x / 3a_0^3 k_B)$  where  $x$  is the Mn concentration, which in our case is  $x \sim 33.3\%$ . For the PM junction we set  $T_C \sim 0K$ .

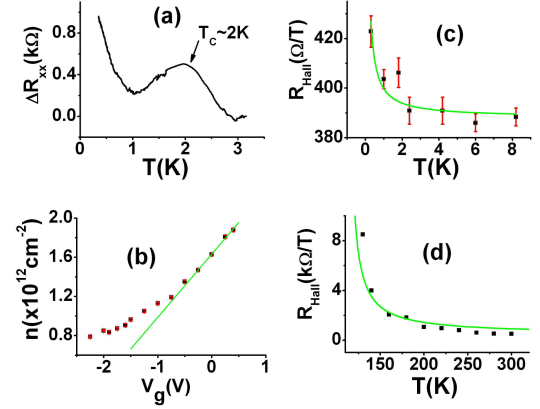


FIG. 4: (a) Longitudinal resistance as a function of temperature gives a Curie temperature value of about 2K. (b) Carrier's density as a function of temperature for a ferromagnetic junction. AHE fitting using (c)  $R_{Hall} = R_0 + C/\mu_0 T$  in our sample and (d)  $R_{Hall} = 450\Omega/T + C/\mu_0(T - 112K)$  in the sample by [10].

The fit shown in figure 4(c) yielded  $R_0 = 388\Omega/T$ , and  $R_S \sim 2.73 \pm 0.58 \times 10^{-11}\Omega/T$  for the AHE coefficient. Comparing to the AHE coefficient  $R_S \sim 4.46 \pm 0.76 \times 10^{-7}\Omega/T$  for a p-type GaMnAs system, extrapolated from the data of Nazmul et al. [10] (figure 4(d)), we find our  $R_S$  value to be  $\sim 16,000$  times smaller. To obtain a more accurate bound, it is necessary to account for the two conducting components in parallel. Since the AHE coefficient is associated only with the electrons residing in the QW, it is their contribution that needs to be evaluated. We begin by expressing the Hall coefficient of the two components in the following form:

$$R_{Hall} = -\frac{n_{QW}\mu_{QW}^2 + n_{HJ}\mu_{HJ}^2}{e(n_{QW}\mu_{QW} + n_{HJ}\mu_{HJ})^2} \quad (2)$$

where  $e$  is the magnitude of the electron charge,  $n_{QW}$  the electron density in the QW,  $n_{HJ}$  the electron density in the HJ and,  $\mu_{QW}$  and  $\mu_{HJ}$  are the respective mobilities. In this sample, a one-component analysis of the density dependence on gate voltage near  $V_g \sim 0V$  signified a depth for the 2DEG that is in between the

depths of the QW and the HJ. (See Fig. 4(b)), indicating that both two channels are contributing. To disentangle the two contributions we note that Eq. 2 indicates that the fractional size of the QW contribution to  $R_{Hall}$  is  $(n_{QW}\mu_{QW}^2)/[n_{QW}\mu_{QW}^2 + n_{HJ}\mu_{HJ}^2] = qr^2/(qr^2 + 1)$ , where  $q = n_{QW}/n_{HJ}$ , and  $r = \mu_{QW}/\mu_{HJ}$ . From Fig. 4(b) the slope for the density versus  $V_g$  at  $V_g = 0$  V, in a 1- component analysis, i.e.  $n \propto (1/R_{Hall})$ , is  $0.75 \pm 0.1 \times 10^{12} \text{cm}^{-2}/\text{V}$ . This compares to an expected slope of  $1.44 \times 10^{12} \text{cm}^{-2}/\text{V}$  for the QW 2DEG at a depth of 50 nm below the surface. This reduced slope by a factor of 1.9 implies a depth of  $\sim 96$  nm, which is at 1.9 times the depth of the QW 2DEG but  $\sim 1/2$  times that for the HJ 2DEG. As  $V_g$  is reduced below 0 V, down to  $V_g = -1.5$  V, the slope gradually decreases before reaching the value expected for the HJ as the QW becomes empty. This difference of 1.5 V indicates that at  $V_g = 0$  V, there is a density of  $\sim 2.15 \times 10^{12} \text{cm}^{-2}$  in the QW. The HJ density at  $V_g \geq -1.5$  V is  $0.85 \times 10^{12} \text{cm}^{-2}$ . At  $V_g = 0$  V, this gives a ratio between the two densities of  $q = n_{QW}/n_{HJ} \sim 2.15/0.85 = 2.53$ . Since  $n_{QW}$  is linearly proportional to the change in  $V_g$  we take the derivative of  $1/R_{Hall}$  with respect to  $n_{QW}$  in Eq. (2) to compare to the experimental slope:

$$\frac{d(1/R_{Hall})}{dn_{QW}} = e \frac{[q^2 r^4 + 2qr^2 + 2r - r^2]}{(qr^2 + 1)^2}. \quad (3)$$

Setting it to  $e/1.9$  at  $V_g = 0$  V, and simultaneously matching the value of the density obtained from  $n \propto 1/R_{Hall}$  using Eq. (2) (1 component), of  $1.6 \pm 0.02 \times 10^{12} \text{cm}^{-2}$ , we find that  $r \sim 0.22$  and  $q = 2.5 \sim 2.15/0.85 = 2.53$ . Finally, compute the factor:  $qr^2/(qr^2 + 1) \sim 1/9$ . We therefore expect a reduction of a factor  $\sim 9$  for the AHE coefficient ( $\sim 16,000$ , yielding a lower bound that is still  $\sim 1,800$  times smaller than the p-type system[10]. Although the mobility  $r$  is on the low side in this crude estimate, it is still roughly of order unity.

The difference in density and mobility of this Hall device, compared to the first device, could arise from inhomogeneity in the Mn atom density across the 2 inch crystal wafer. Although the wafer was rotated during growth, at the approximate speed of 1 revolution every 2 seconds, it takes less than 1 second to deposit a monolayer. Thus the difference in Mn atom flux across the wafer is accentuated. Since Mn acts as effective acceptors, at  $2/3$  ML, even a 1 percent variation in flux could account for the observed differences.

Our results indicate that the AHE is either absent or at least very small compared to an equivalent system where the carrier is the hole. This seems to confirm the predictions made by Jungwirth et al. in [7], where they show that in the case of a system with small or no spin-orbit interaction, the AHE coefficient should be very small compared to a system with large spin-orbit interaction.

As a final note, we speculate on the nature of the ferromagnetic interaction. In p-type carrier mediated ferromagnetic GaMnAs systems, the magnetic interaction is believed to be of the Zener type, where double-exchange through the hole-impurity band gives rise to the magnetic coupling. Despite the complexity of our structure with both Si (n-type) and Be (p-type) dopants, all in the presence of Mn atoms, which act themselves as acceptors, the fact that the GaMnAs QW can be filled with electrons indicates that all acceptor levels are filled. Therefore, hole-mediation cannot take place. For the QW 2DEG in our first sample, the mobility and the density ( $\mu \sim 598 \text{cm}^2/\text{Vs}$  and  $n \sim 0.72 \times 10^{12} \text{cm}^{-2}$ ) yield a mean free path (mfp) and Fermi wavelength of  $l_{mfp} \sim 8.30 \text{nm}$  and  $\lambda_F \sim 29.5 \text{nm}$ , respectively. Since  $l_{mfp}$  is significantly smaller than the  $\lambda_F$ , it is quite likely that conduction could occur through an impurity band as well. Moreover, some form of anisotropy in the exchange interaction must be in place to stabilize the 2-d magnetism, such as the dipolar interaction with an energy scale of  $\sim 100 \mu\text{eV}$ .

We would like to thank Qian Niu for useful discussions. Angelo Bove would like to thank Ndeye Khady Bove for her support. This work was supported by NSF DMR-0135931.

---

\* Electronic address: yingshe@phy.duke.edu

- [1] H. Ohno, Science **281**, 951 (1998).
- [2] Ohno, H., H. Munekata, T. Penney, S. von Molnr, and L. L. Chang, Phys. Rev. Lett. **68**, 2664 (1992).
- [3] G. T. Thaler, M. E. Overberg, B. Gila, R. Frazier, C. R. Abernathy, S. J. Pearton J. S. Lee, S. Y. Lee, Y. D. Park, Z. G. Khim, J. Kim and F. Ren, Appl. Phys. Lett. **80**, 3964 (2002).
- [4] Kwang-Soo Huh, Moon-Ho Ham, Jae-Min Myoung, Jeung-Mi Lee, Kyoung-Il Lee, Joon-Yeon Chang, Suk-Hee Han, Hi-Jung Kim and Woo-Young Lee, Jpn. J. Appl. Phys **41**, 1069 (2002).
- [5] X.H. Xu, A.J. Behan, M. Ziese, H.J. Blythe, J.R. Neal, A. Mokhtari, M.R. Ibrahim, A.M. Fox and G. A. Gehring, New Journal of Physics **8**, 135 (2006).
- [6] A. Bove, F. Altomare, N. B. Kundtz, A. M. Chang, Y. J. Cho, X. Liu and J. Furdyna, arXiv:0802.3863v1 [cond-mat.mes-hall].
- [7] T. Jungwirth, Qian Niu and A. H. MacDonald, Phys. Rev. Lett. **88**, 207208 (2002).
- [8] T. Jungwirth, J. Masek, Jairo Sinova, J. Kucera and A.H. MacDonald, Rev. Mod. Phys. **78**, 809 (2006).
- [9] J. K. Furdyna, J. Appl. Phys **64**, R29 (1988).
- [10] A. M. Nazmul, S. Sugahara, and M. Tanaka, Phys. Rev. B **67**, 241308(R) (2003).
- [11] A. M. Nazmul, T. Amemiya, Y. Shuto, S. Sugahara and M. Tanaka, Phys. Rev. Lett. **95**, 017201 (2005).
- [12] D. J. Priour, E. H. Hwang, and S. Das Sarma, Phys. Rev. Lett. **95**, 037201 (2005).